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Experimental studies of the absorption and emissions from laser-induced spark in combustible gases

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Abstract

This paper reports preliminary results of experimental measurements of the emission and absorption of laser-induced spark in air, O_2, N_2, H_2, and CH_4 using a Q-switched Nd-Yag laser operating at 532 nm and 1064 nm, with a pulse duration of 5.5 ns. Line radiation spectra observed for spark in air consist of N and O lines that are distinctly observed in the region from 399.49 to 870.2 nm. The spectra of spark created in CH_4 show strong H lines centered at 656.46 nm. Some rather weak emission lines are also observed at 460.4, 497.0, 497.3, and 503.7 nm. Laser wavelength has a strong effect on the absorption data obtained as a function of total laser energy E_o. However, in case of variable pressure, its effect is considered to be within experimental fluctuations. Published by Elsevier Science B.V.

1. Introduction

Interest in laser ignition has increased in recent years because of its many potential benefits over conventional ignition systems. In general, laser ignition is a non-intrusive technique and is capable of providing multiple ignition sites that can be programmed to ignite a gaseous combustible mixture either sequentially or simultaneously. Use of multipoint ignition, a flame can be initiated simultaneously at many points throughout the mixture volume, the total burning time could be much smaller [1]. This could be potentially important for fuel-lean combustion and for high-speed combustion applications.

Laser spark ignition studies [2–5] have shown that, when a powerful laser beam interacts with a combustion mixture, a spark plasma of high temperature and high pressure is created. This extreme condition relative to the ambient gas leads to the development of a rapidly expanding shock wave that is of sufficient strength to ignite flammable mixtures. Thus, for better understanding of laser spark ignition it is important to study the physical and chemical processes underlying the formation, evolution, and interaction between the spark and its surrounding combustible gases. Such an understanding requires a knowledge of many fundamental properties of the spark including its absorption coefficient. In addition, spark produced this way has a large concentration of electrons, ions, and other excited species. Since these excited species affect strongly the chemical ignition process, quantifying the existence of

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these species is also critical. In our previous paper [6] we have reported breakdown threshold intensities for many common combustion gases. This present work will focus on the measurements of laser-induced spark emissions and absorption. The results are presented in the next sections.

2. Experimental apparatus

The experimental apparatus used in this study was described by Phuoc and White [2]. In brief, it consisted of a single-mode, Q-switched Nd-Yag lasers (Quantel, Brilliant W) which was operated at either 1064 nm or 532 nm, with pulse duration of 5.5 ns. The laser beam was delivered and focused into the ignition cell using a 75 mm focal lens and a 1% to 99% variable beam splitter. With this laser system, the laser spot diameter was estimated to be 17 μm for a 1064 nm beam and 8.5 μm for a 532 nm beam. In order to have a well controlled beam energy throughout the experiments, each laser potential controller was set at a fixed location, and the laser energy from the laser was varied by rotating the beam splitter about its center. The experimental gases — methane, hydrogen, nitrogen, oxygen (Matheson, research grade with 99.99% purity) — and air from high-pressure cylinders were delivered to the ignition cell using a gas handling system with precise gas flow controllers.

For space-resolved studies of laser-induced spark emission, the emissions lines were monitored using a Multichannel Instaspec IV CCD detection system (Oriel). It consisted of a front illuminated 1024 × 128 pixel format CCD detector head (Model 78430, Oriel), a MS260i imaging spectrograph (Model 74050, Oriel), a single-track fiber optic cable (Model 77403, Oriel), and a F-NO matcher (Model 77529, Oriel). The MS260i had a micrometer driven entrance slit with its width is variable from 4 μm to 3 mm and its height is from 2 to 12 mm. It was equipped with three gratings covering a spectrum of 250 to 900 nm. The emission light from the spark plasma was collected using the fiber-SMA with the F-NO matcher attached to the entrance slit of the MS260i spectrograph which was set at 25 μm for the present study. The spectral lines spread over through the MS260i exit slit and were detected by the detector which was operated under full-vertical binning mode.

Two pyroelectric energy meters detectors (Oriel 70713), together with two energy readout units (Oriel 70833) were used for spark energy measurements. One meter was placed behind the exit window facing the incoming laser beam, and the other was placed after the variable beam splitter which was located before the entrance window. If the energy loss due the exit window is known, this arrangement allowed the energy meters to detect the transmitted beam through the test cell with and without breakdown. Let E_o be the laser energy delivered to the experimental cell, E_{abs} be the energy absorbed by the spark, and E_m be the laser energy measured by the detector behind the exit window the transmitted laser energy through the spark, E_{tr}, is corrected as follows

$$E_m = (E_o - E_{abs})(1-x), \tag{1}$$

$$E_{tr} = E_o - E_{abs} = \frac{E_m}{1-x}, \tag{2}$$

where x is the portion by which the laser energy is lost due to the exit window. To determine x laser energy less than the threshold energy for gas breakdown was fired through the experimental cell and the transmitted beam with and without the exit window was measured. We found that x fluctuated between 5 and 10%. For the present work we approximated x at 7.5% for all calculations.

3. Results and discussions

Visible observation showed that laser-induced sparks in air, nitrogen, and oxygen were very bright, while those produced in methane and hydrogen had a pinkish color. Our present spectroscopic measurements indicated that there were two main contributions to the emitted spectrum: the spectrally broad band background continuum due to the luminous plasma, and the line radiation due to the ionized gas species. The continuum contribution was found to be significant during the early stage of the spark development, while the contribution of the line radiation was dominant during the plasma cooling stage. Fig. 1 shows typical space-resolved spectra of a laser-spark created in air, nitrogen, oxygen and methane.

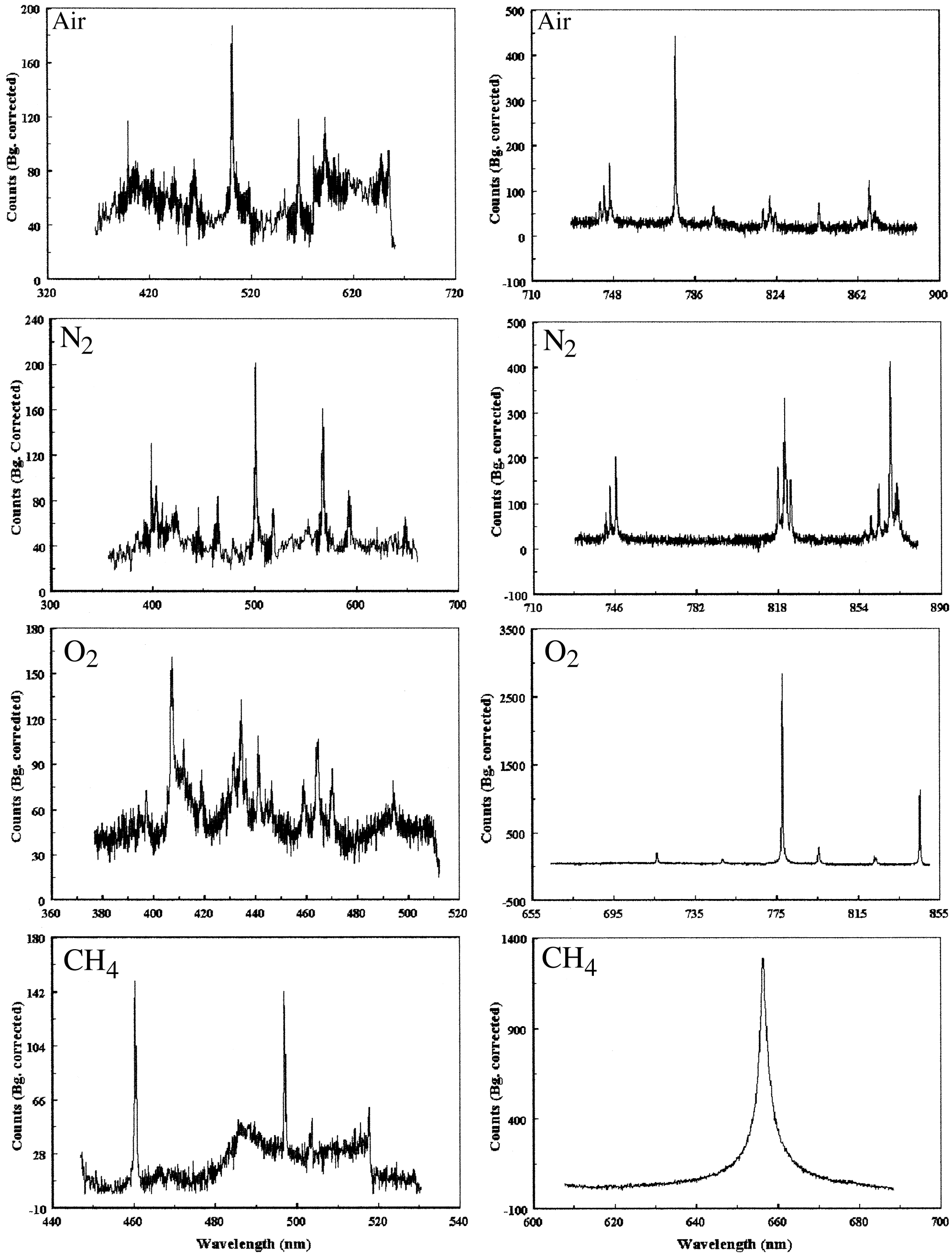

Fig. 1. Space-resolved spectra of laser-induced spark created in air, N_2, O_2, and CH_4 (760 torr; $E_o = 18.5$ mJ).

In general, all the spectra reported here show a similar behavior. Line radiation intensities in the wavelength region of less than about 600 nm were weak and close to the plasma continuum background. Emissions from lines longer than this region, however, were distinct and strong. The spectra of spark created in CH_4 showed a strong H line centered at 656.46 nm. Some rather weak emission lines were also observed at 460.4, 497.0, 497.3, and 503.7 nm. By comparing the spectra for air with those for

N_2 and O_2, it is clear that line radiation from spark created in air consisted mainly of line emissions from N and O which were spreading from 399.49 to 870.2 nm. For N emission lines, strong emissions were observed at around 745.9 to 748.89, 819.0 to 823.78, 869.99 to 871.35 nm while emissions at 399.49, 409.8 to 410.99, 422.9, 500.2 to 503.3, 566.66 to 567.99 nm were weak and close to the plasma continuum background. For O emission lines, line radiation at 716.2, 717.2, 777.19, 777.3, 777.6, 780.7, 795.4, 823.6, 845.3 nm and in the region from 407.4 to 464.5 nm were observed. The strongest line intensities were seen at 777.19, 777.3, 777.6 nm while the intensities of lines in the region from 407.4 to 464.5 nm were seen to be close to the plasma continuum background.

Fig. 2 show the ratio of transmitted laser energy to the total laser energy (E_{tr}/E_o). By using the arrangement for the two pyroelectric energy meters described previously. The transmitted laser energies through the focal volume, with and without breakdown, were measured in the following manner. First, the cell was evacuated to about 10 torr or less, the laser was fired, and the laser energy transmitted through the focal volume was recorded. The cell was then filled with a test gas and the laser was fired again. The spark energy was determined by comparing the energy transmitted through the cell in the presence of breakdown against that in the absence of breakdown. The attenuation of the laser energy in the presence of breakdown is attributed to spark absorption and the scatter of the laser light by the developing spark plasma. However, as discussed by Syage et al. [5], losses due to diffraction are negligible, and such an attenuation can be considered to be due solely to the absorption by the spark. We also neglected the contribution of the reflected beam from the exit window that might add additional energy into the spark column. This is because: (i) all the optics were anti-reflection coated; and (ii) our data show that the laser energy reaching to the exit window was small compared to that absorbed by the spark. If there is any reflection, such reflected energy would be insignificant. In addition, it is noticed that for this present study the exit window was about 31 mm behind the spark (focal spot) and the focal length was 75 mm. Thus, the reflected beam from the exit window would expand and have a diameter of about 6 mm (the original beam diameter) when it reaches the spark whose average diameter is about 0.4–1.2 mm [2]. Thus, the spark can intercept only about less than 3% of the reflected energy. This is too small.

The curves in Fig. 2 show that when pressure was below 50 torr or E_o was below its threshold level all the present test gases were virtually transparent at the experimental laser wavelengths, and the laser energy was transmitted through the focal volume without attenuation. When either E_o or pressure increased E_{tr}/E_o decreased drastically. This is because increasing either E_o or pressure will lead to gas breakdown, a spark plasma is generated, and attenuates the laser energy strongly primarily due to the electron–ion inverse bremsstrahlung process, in which light is absorbed as a result of free–free transitions of the electrons in the field of the ions. The results also show a similar pattern of the effect of the wavelength on E_{tr}/E_o for all the test gases. It is clear that, in case of increasing pressure, the effect of the laser wavelength on E_{tr}/E_o can be considered to be within the limit of experimental fluctuations. However, for the case of increasing E_o, such an effect became more profound. For example, for air, E_{tr}/E_o measured at 1064 nm remained constant at about 1 for E_o up to about 7 mJ while E_{tr}/E_o obtained at 532 nm started to decrease as early as $E_o = 3$ mJ and it dropped drastically to about 50% when E_o was about 7 mJ. Such a significant effect might be due to the fact that the multiphoton ionization process may play a more important role in the spark development at $\lambda = 532$ nm than at $\lambda = 1064$ nm [6].

We assume that the absorption coefficients is calculated using the Beer–Lambert relation:

$$\frac{E_{tr}}{E_o} = \exp(-K_v l), \tag{3}$$

where K_v is the absorption coefficient (1/cm), and l is the length of the focal volume (cm) which can be estimated by the following equation:

$$l = \left(\sqrt{\pi} - 1\right)\frac{\theta}{d} f^2, \tag{4}$$

where f is the focal length, d is the beam diameter, and θ is the beam divergence. For the present laser

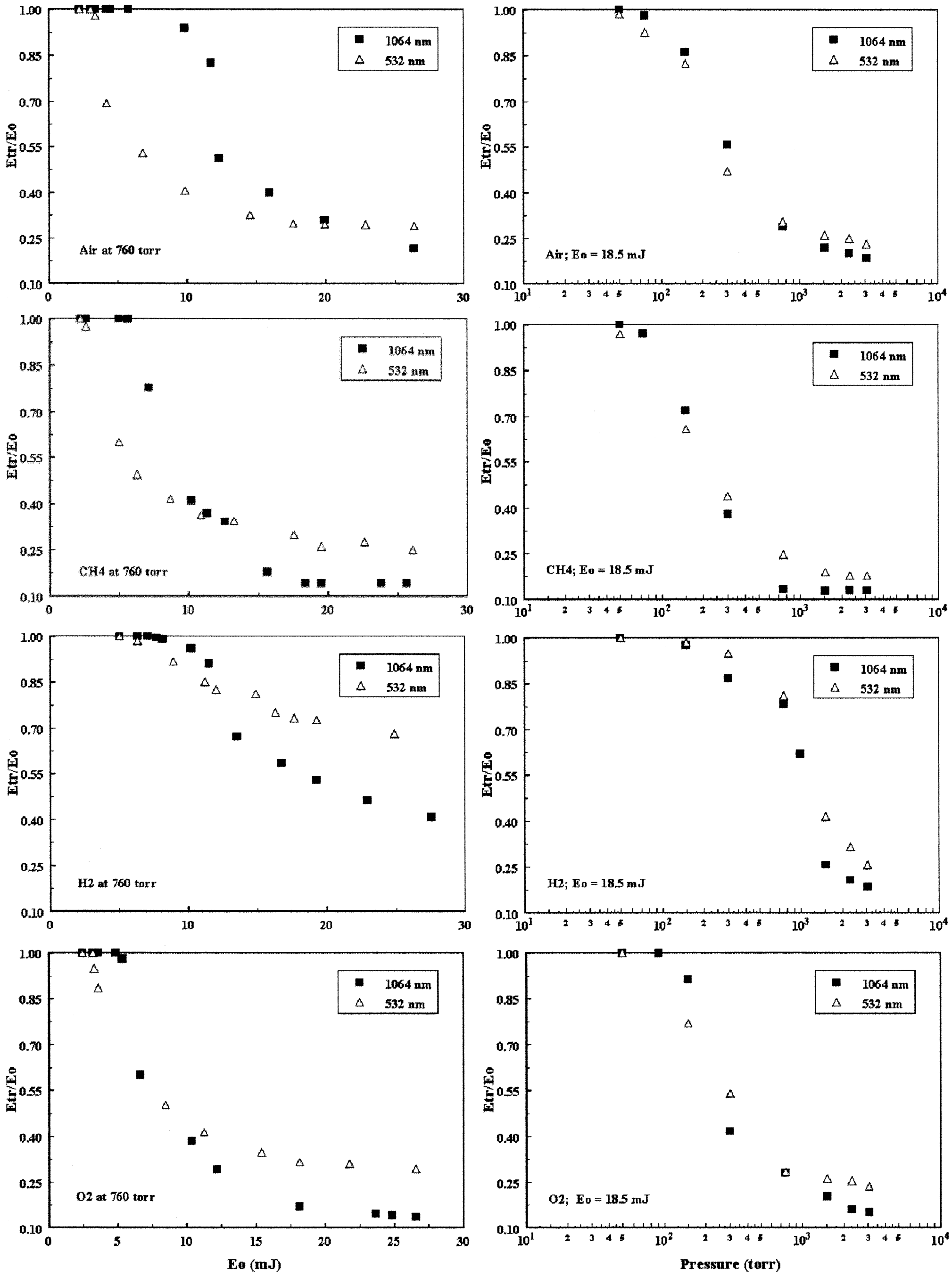

Fig. 2. Ratio of the energy transmitted through the spark to the total laser energy E_{tr}/E_o as a function of E_o or pressure.

condition: $f = 7.5$ cm, $\theta = 0.5$ mrad, and $d = 0.6$ cm we estimated the focal $l = 194$ μm. The results for K_v are shown in Fig. 3 for sparks created in air, CH_4, H_2, and O_2. As E_o, or pressure increased, K_v increased dramatically in the region where E_o was less than about 12 mJ or pressure was less than about

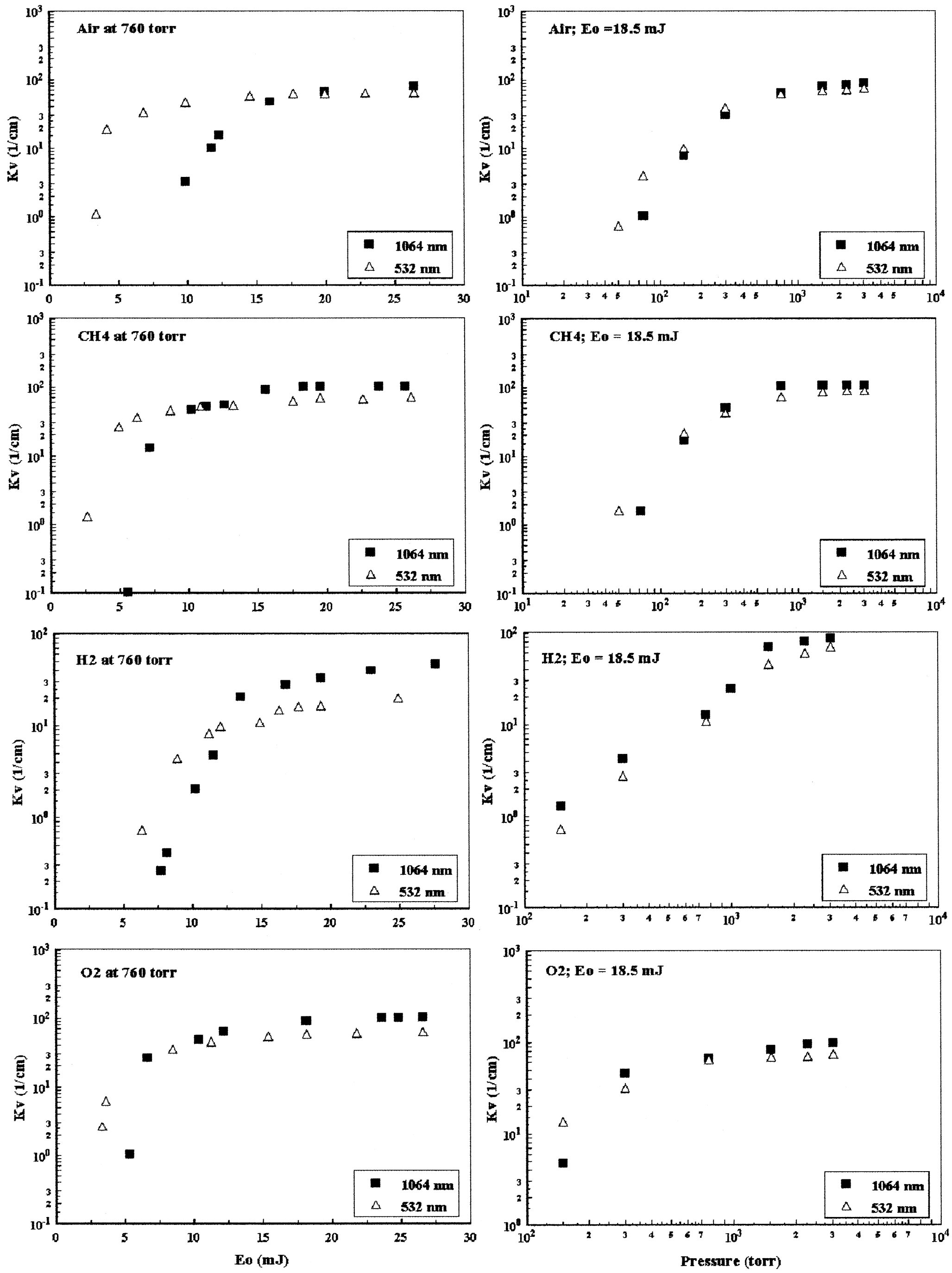

Fig. 3. Absorption coefficient of laser spark in air, CH_4, H_2, and O_2 as a function of laser energy E_o and pressure.

760 torr and it leveled off at about 75 to 89 cm^{-1} at higher E_o or higher pressure. The results for air and CH_4 reported here are in agreement with the absorption coefficients reported in literature [7].

4. Conclusions

Emission and absorption of a laser-induced spark in common combustion gases were measured. Laser sparks were created using a Q-switched Nd:Yag laser operating at 1064 nm and 532 nm with pulse duration of 5.5 ns. Line radiation spectra observed for spark in air consisted of N and O lines while spark created in CH_4 emitted strong H lines centered at 656.46 nm. Absorption data showed a strong dependence on both laser energy and pressure. Using Beer–Lambert relation the absorption coefficient K_v was calculated. For spark created in air, K_v was found to be in agreement data reported in literature [7].

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